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(54) Title: PESTICIDAL COMPOSITIONS

(57) Abstract

The use in a pesticidal composition of at least one aliphatic mono-, di- or triester, optionally substituted by one or more hydroxy groups, said ester(s) having: i) a log octanol-water partition coefficient (log P) of from 5 to 13.1, ii) an equivalent hydrocarbon (EH) value of form 20 to 42; the EH value being calculated as the number of carbon atoms plus five for any hydroxy group present plus 3 for each ester group, and iii) an alcohol moiety comprising at least 2 carbon atoms.

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Pesticidal compositions

Field of the invention

This invention concerns pesticidal compositions.

Background to the invention

Pesticidal compounds, particularly those used for crop protection, are commonly used in the form of compositions containing one or more co-formulants, for example surfactants. We have found that a new group of compounds, not previously used in association with pesticidal compounds, can also be used with advantage in association with pesticidal compounds.

Description

In one aspect, the invention provides the use in a pesticidal composition of at least one aliphatic mono-, di- or triester, optionally substituted by one or more hydroxy groups, said ester(s) having:

i) a log octanol-water partition coefficient (log P) of from 5 to 13.1,

ii) an equivalent hydrocarbon (EH) value of from 20 to 42; the EH value being calculated as the number of carbon atoms plus five for any hydroxy group present plus 3 for each ester group, and

iii) an alcohol moiety comprising at least 2 carbon atoms.

The term 'ester of the invention' is used herein to indicate an ester having the above properties.

In another aspect the invention provides a pesticidal composition which comprises a pesticidal compound and at least one ester of the invention.

In a further aspect the invention provides a method of combating pests at a locus infested or liable to be infested therewith which comprises applying to said locus an effective amount of a pesticidal composition

which comprises a pesticidal compound and at least one ester of the invention.

The log P value of an ester as that term is used herein is that calculated from the structure of the compound using the clog P Program, Pomona College Med Chem release 3.54, January 1988, provided by Daylight Chemical Information Systems Inc, Claremont, California. The values generated by this program correlate well in general with those determined experimentally by methods well known to those skilled in the art, but such methods are difficult to apply accurately to compounds having log P values within the scope of this invention, and we therefore use the calculated values.

The EH value can be simply calculated from the structure of each compound. Sometimes, however, the esters of the invention occur or are presented as mixtures of individual compounds, and the mixture will therefore include compounds of different EH values.

The ratio of EH value to log P value is preferably in the range 2.4:1 to 4.15:1.

Preferred esters of the invention are succinate diesters, lactic acid carboxylic esters, and esters resulting from esterification of a saturated acid of at least 12 carbon atoms with a branched chain alcohol.

Examples of suitable esters of the invention include propylene glycol di-ester of coconut fatty acids (C8 ester log P=7.022, EH=25; C10 ester log P=9.138, EH=29), isopropyl palmitate (log P=8.386, EH=22), isopropyl myristate (log P=7.328, EH=20), 2-ethylhexyl 12-hydroxystearate (log P=9.882, EH=34), dioctyl succinate (log P=7.771, EH=26), 2-ethylhexyl palmitate (log P=11.03, EH=27), myristyl lactate (log P=6.556, EH=25), lauryl lactate (log P=5.598, EH=23), and triglycerides of C_8-C_{10}-coconut fatty acids (C8 ester log

P=9.89, EH=36; C10 ester log P=13.064, EH=42).

We have found that the esters of the invention, which have little or no pesticidal activity in their own right, surprisingly aid the penetration of the pesticidal compound into plant or pest tissue, thereby promoting enhanced activity, particularly of pesticidal compounds whose activity depends on such penetration.

The pesticidal compound is especially one disclosed in The Pesticide Manual, 10th edition, published by Crop Protection Publications, and may for example be a herbicide, fungicide, insecticide or acaricide.

It is preferably a phytopathogenic fungicide selected from:

(i) a conazole steroid demethylation inhibitor;

(ii) a steroid reduction inhibitor based on a 1-[3-(4-tert-butylphenyl)-2-methylpropyl] group which is attached via the N-atom to piperidine or 2,6-dimethylmorpholine

(iii) a dithiocarbamate fungicide

(iv) a phthalimide fungicide in which a chloroalkylthio group is attached via the N-atom to the optionally hydrogenated phthalimide group.

(v) an anilide fungicide

(vi) an mbc fungicide.

(vii) a carbamate fungicide

(viii) a copper compound fungicide

(ix) a tin compound fungicide

(x) a strobilurin type fungicide,

(xi) a 2-anilinopyrimidine fungicide or

(xii) a fungicide selected from the group consisting of chlorothalonil, dimethomorph, fenpiclonil, fluazinam, hymexazol, nuarimol, pencycuron, pyrifenox, thicyofen, probenazole, pyroquilon, tricyclazole, quaternary ammonium compounds,

fludioxonil, 5-chloro-2-methyl-4-isothiazolin-3-one, 2-methyl-4-isothiazolin-3-one (and mixtures of these two), furmecyclox, 3-iodo-2-propynyl butylcarbamate and sulfur.

Conazoles are as defined in ISO standard 257, ie compounds based on imidazole or 1,2,4-triazole and containing a halogenated phenyl group. Examples include prochloraz (and its metal complexes, especially the manganese or copper complex), propiconazole, flusilazole, hexaconazole, tebuconazole, difenoconazole, bromuconazole, cyproconazole, diniconazole, fenbuconazole, imibenconazole, furconazole, tetraconazole, myclobutanil, penconazole, fluquinconazole, azaconazole, imazalil, triflumizole, epoxiconazole, triticonazole, metconazole and the fungicide having the code No SSF 109.

Examples of type (ii) fungicides include fenpropimorph and fenpropidin.

Examples of type (iii) fungicides include mancozeb and thiram.

Examples of type (iv) fungicides include folpet, captafol and captan.

Examples of type (v) fungicides include

a) 3',5'-dichloroanilide fungicides in which the anilino nitrogen comprises a ring carrying two oxo substituents, in positions adjacent the nitrogen, eg iprodione, vinclozolin or procymidone, or
b) acetanilide fungicides, eg metalaxyl or ofurace,
c) sulfanilide fungicides, eg dichlofluanid,
d) benzanilide fungicides, eg flutolanil, and
e) heteroarylanilide fungicides, eg thifluzamide.

Examples of type (vi) fungicides include carbendazim, benomyl and thiophanate-methyl.

Examples of type (vii) fungicides include

diethofencarb and propamocarb.

Examples of type (viii) fungicides include Bordeaux mixture, oxine-copper, copper oxychloride and copper naphthenate.

Examples of type (ix) fungicides include tributyltin oxide and tributyltin naphthenate.

Strobilurine type fungicides (type (x) fungicides) are methyl esters of arylacetic acid in which the acetic acid also carries a methoxymethylene or methoxyimino substituent. The aryl group is usually a 2-substituted phenyl group. Examples of such compounds are those disclosed in a wide number of patent applications, including EPs 178826, 203606, 203608, 206523, 229974, 226917, 242070, 242081, 243012, 243014, 251082, 256667, 260794, 260832, 267734, 270252, 273572, 274825, 278595, 291196, 299694, 307101, 307103, 310954, 312221, 312243, 329011 and 336211. Specific compounds are those having the code Nos BAS 490F and ICIA 5504.

Examples of type (xi) fungicides include pyrimethanil, mepanipyrim and cyprodinil.

Examples of insecticides which may be used include amitraz, triazophos and formetanate.

Examples of acaricides which may be used include clofentezine.

The names quoted for these compounds are the non-proprietary common names and the chemical structures can be found for example by reference to The Pesticide Manual.

The invention is particularly useful for pesticidal compounds of high melting point and/or low solubility in organic solvents. It is particularly applicable to fluquinconazole.

The esters of the invention may be incorporated in conventional concentrate formulations of the pesticidal

compound (eg suspension concentrates, suspoemulsions or solid formulation types such as water dispersible granules), which are diluted with water prior to application, or they may be added to the pesticidal composition just prior to use.

It may be necessary and/or desirable for the pesticidal composition to include small quantities of solvent and/or surfactant, especially a non-ionic surfactant, and other additives such as fatty acids, to improve the emulsifiability of the esters of the invention. Such modifications are well within the competence of those skilled in the art.

The esters of the invention preferably comprise from 2 to 90% by weight of a concentrate formulation.

The esters of the invention are preferably applied in a diluted formulation at a concentration of 20g to 2000g, preferably 50g to 500g, per 100 litres.

The mixtures can be used for those applications where the pesticide would normally be used. In the case of fluquinconazole, for example, it is preferably used in the direct treatment of growing crops, such as cereals and fruit, or in seed treatments.

The following Examples illustrate the use of esters of the invention, in which:

"Crodamol OHS" is a trade name for 2-ethylhexyl 12-hydroxystearate;

"Crodamol PC" is a trade name for propylene glycol diester of coconut fatty acids;

"Crodamol IPP" is a trade name for isopropyl palmitate;

"Crodamol IPM" is a trade name for isopropyl myristate;

"Crodamol OHS" is a trade name for 2-ethylhexyl 12-hydroxystearate;

"Crodamol OSU" is a trade name for dioctyl succinate;

"Crodamol OP" is a trade name for 2-ethylhexyl palmitate;

"Crodamol ML" is a trade name for myristyl lactate;

"Crodamol LL" is a trade name for lauryl lactate.

Example 1

In a glasshouse wheat plants were inoculated with *Erysiphe graminis*. Three days after inoculation the plants were sprayed separately with:

(a) fluquinconazole (FQ), obtained by diluting with water to the desired concentration, a wettable granule comprising 25% by weight active ingredient,

(b) Crodamol OHS, or

(c) Crodamol PC,

and with a mixture of (a) with (b) or (c) containing about 10% of an emulsifier.

8 replicates were used for each treatment. After 4 days, the plants were assessed for the control of sporulation of the disease achieved by the treatments.

The individual components (a), (b) and (c) alone had no effect on sporulation.

Results for the mixtures are given in the following table:

AI	g/ha	(b) (%v/v)	(c) (%v/v)	% control of sporulation
FQ	100	0.4		95.8
FQ	100	0.2		80
FQ	100		0.4	96.9
FQ	100		0.2	89.2

Example 2

In a glasshouse wheat plants were inoculated with *Erysiphe graminis*. One day after inoculation the plants were sprayed with:

(a) an active ingredient (AI), alone and with

(b) Crodamol OHS, or

(c) Crodamol PC.

8 replicates were used for each treatment, and each treatment was made by an automatic sprayer delivering the equivalent of 200 l/ha of formulation. After 7 days, the plants were assessed for the control of the disease achieved by the treatments.

Results are given in the following table, in which:

EC is epoxiconazole

BC is bromuconazole

TC is tebuconazole

FP is fenpropimorph

PM is pyrimethanil

CP is cyprodinil.

AI	g/ha	(b) (%v/v)	(c) (%v/v)	% control
EC	31.25	0.4		97.9
EC	31.25	0.2		98.2
EC	31.25		0.4	94.8
EC	31.25		0.2	98.2

	EC	31.25			82.6
	BC	62.5	0.4		94.6
	BC	62.5	0.2		89.4
	BC	62.5		0.4	94.8
	BC	62.5		0.2	96.1
	BC	62.5			77.9
	TC	31.25	0.4		96.4
	TC	31.25	0.2		96.1
	TC	31.25		0.4	94.6
	TC	31.25		0.2	95.8
	TC	31.25			87.8
	FP	93.75	0.4		62.1
	FP	93.75	0.2		73.0
	FP	93.75		0.4	75.1
	FP	93.75		0.2	67.8
	FP	93.75			10.1
	PM	800	0.4		54.3
	PM	800	0.2		51.9
	PM	800		0.4	56.8
	PM	800		0.2	52.2
	PM	800			28.9
	CP	375	0.4		100
	CP	375	0.2		100
	CP	375		0.4	98.4
	CP	375		0.2	100
	CP	375			87.0
	Untreated Control				0

Example 3

Two week old bean plants (Vicia faba) infested with Aphis fabae were sprayed at a volume equivalent to 1000 l/ha with

(a) an active ingredient (AI), alone and with

(b) Crodamol OHS,

as set out in the table below.

After drying, the plants were placed in a greenhouse, and were assessed 3 days later relative to an untreated control.

Results are given in the following table, in which:

AZ is amitraz

TP is triazophos

FM is formetanate.

AI	g/ha	(b) (%v/v)	% Mortality
AZ	300	-	0
AZ	300	0.3	50
AZ	100	-	0
AZ	100	0.3	23
AZ	30	-	0
AZ	30	0.3	10
TP	300	-	35
TP	300	0.3	65
TP	100	-	0
TP	100	0.3	45
TP	30	-	0
TP	30	0.3	35
FM	300	-	0
FM	300	0.3	25

Example 4

Four week old bean plants (Phaseolus vulgaris) were infested with Tetranychus urticae. After 1 day and after 2 days, the motile stages were blown away, and the plants and remaining eggs were sprayed by hand to runoff with

(a) clofentezine (CZ) (3g/hl) alone,

(b) 0.3% of the esters of the invention specified below, alone, and

(c) mixtures of (a) and (b).

After drying, the plants were placed in a greenhouse. 10 days later, the plants were assessed for

ovicidal activity relative to an untreated control

Results are given in the following table:

Ester	% Mortality with CZ	% Mortality without CZ
Crodamol IPP	99	0
Crodamol IPM	90	0
Crodamol OP	98	8
Crodamol LL	97	0
Crodamol PC	63	0
Crodamol OHS	80	15
None	50	0

Example 5

The penetration efficacy of various esters of the invention was demonstrated by applying an acetone/water solution of radio-labelled fluquinconazole (0.05% w/v) both alone and containing the esters specified below to vine plants using a microapplicator. After 5 days, the fluquinconazole remaining on the surface of the plants was recovered and determined quantitatively, the difference between the percentage remianing and 100% being indicative of the amount of compound penetrating the plant tissue. Results are given in the following table:

Ester	%w/v	Surface %
Crodamol PC	0.5	27
Crodamol IPP	0.5	5
Crodamol IPM	0.5	10
Crodamol OHS	0.5	19
Crodamol OSU	0.5	28
Crodamol OP	0.5	26
Crodamol ML	0.5	10
Crodamol LL	0.5	22
None	-	>90

CLAIMS

1. The use in a pesticidal composition of at least one aliphatic mono-, di- or triester, optionally substituted by one or more hydroxy groups, said ester(s) having:
   i) a log octanol-water partition coefficient (log P) of from 5 to 13.1,
   ii) an equivalent hydrocarbon (EH) value of from 20 to 42; the EH value being calculated as the number of carbon atoms plus five for any hydroxy group present plus 3 for each ester group, and
   iii) an alcohol moiety comprising at least 2 carbon atoms.
2. The use according to claim 1 of at least one ester having a ratio of EH value to log P value in the range 2.4:1 to 4.15:1.
3. The use according to claim 1 or claim 2 of at least one ester which is a succinate diester, a lactic acid carboxylic ester, or an ester resulting from esterification of a saturated acid of at least 12 carbon atoms with a branched chain alcohol.
4. The use according to any of claims 1 to 3 of at least one ester selected from propylene glycol di-ester of coconut fatty acids, isopropyl palmitate, isopropyl myristate, 2-ethylhexyl 12-hydroxy-stearate, dioctyl succinate, 2-ethylhexyl palmitate, myristyl lactate, lauryl lactate, and triglycerides of C_8-C_{10}-coconut fatty acids.
5. The use according to any of claims 1 to 4 in which the pesticidal composition is a phytopathogenic fungicidal composition.

6. The use according to claim 5 in which the composition comprises fluquinconazole as active ingredient.

7. A pesticidal composition which comprises a pesticidal compound and at least one ester as defined in any of claims 1 to 4.

8. A composition according to claim 7 which comprises at least one ester as defined in any of claims 1 to 4 in a concentration of from 2 to 90% by weight.

9. A method of combating pests at a locus infested or liable to be infested therewith which comprises applying to said locus an effective amount of a pesticidal composition which comprises a pesticidal compound and at least one ester of the invention as defined in any of claims 1 to 4.

10. A method according to claim 9 in which the ester of the invention is applied in a concentration of from 20g to 2000g per 100 litres of the applied composition.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 96/00087

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 A01N37/36 A01N37/12 A01N37/04 A01N37/02 //(A01N37/36, 57:16,47:22,43:84,43:713,43:653,43:54,37:52),(A01N37/12,57:16, 47:22,43:84,43:713,43:653,43:54,37:22),(A01N37/04,57:16,47:22,

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B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
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C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Week 7528 Derwent Publications Ltd., London, GB; AN 75-46209W [28] XP002003965 & DD,A,112 891 (HOECHST) , 12 May 1975 see abstract	1-5,7-10
X	DE,A,23 03 757 (HOECHST AG) 15 August 1974 see page 2, paragraph 2 see page 3, paragraph 2 see page 3, paragraph 4 see page 4, paragraph 4	1-5,7-10
X	DE,A,42 39 181 (HOECHST AG) 5 May 1994 see page 2, line 17 - line 24 see page 3, line 1 see page 3, line 9 - line 10	1-4,7-10

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Date of the actual completion of the international search	Date of mailing of the international search report
28 May 1996	11.06.96

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 96/00087

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 43:84,43:713,43:653,43:54,37:52)

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Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

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C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 5, no. 160 (C-075), 15 October 1981 & JP,A,56 092207 (NISSHIN OIL MILLS), 25 July 1981, see abstract ---	1-4,7-10
X	DE,A,32 47 050 (BAYER AG) 20 June 1984 see page 11, paragraph 4 - page 12, paragraph 1 see page 18, line 12 - line 13 see page 18, line 15 - page 19, line 11	1-4,7-10
Y	see page 11, paragraph 2 ---	4
Y	FR,A,2 347 052 (BAYER AG) 4 November 1977 see page 3, line 6 - page 4, line 21 see page 5, line 33 - page 6, line 16 --- -/--	4

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP,A,0 295 411 (SANSHO CO ;EISAI CO LTD (JP)) 21 December 1988 see page 2, line 30 - line 34 see page 2, line 53 - page 3, line 1	4
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